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A heat pump at a molecular scale controlled by a mechanical force

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Abstract. – We show that a mesoscopic system such as Feynman's ratchet may operate as a heat pump, and clarify a underlying physical picture. We consider a system of a particle moving along an asymmetric periodic structure. When put into a contact with two distinct heat baths of equal temperature, the system transfers heat between two baths as the particle is dragged. We examine Onsager relation for the heat flow and the particle flow, and show that the reciprocity coefficient is a product of the characteristic heat and the diffusion constant of the particle. The characteristic heat is the heat transfer between the baths associated with a barrier-overcoming process. Because of the correlation between the heat flow and the particle flow, the system can work as a heat pump when the particle is dragged. This pump is particularly effective at molecular scales where the energy barrier is of the order of the thermal energy.

Introduction. – In the last decade, physics and technology have moved more and more toward the manipulation of mesoscopic objects. Techniques of micro manipulation of molecules are expanding to open new possibilities to future technology. Although to control heat flow in mesoscopic scales may be among crucial issues, its study is still in a preliminary stage [1]. Heat pumps working in nano-scale devices [2, 3] or mesoscopic machines [4] are studied but the number of cases are still limited. In this Letter, we study a class of mesoscopic systems working as a heat pump and present a physical picture explaining how they work. We start from a numerical study adopting one specific model, and demonstrate that a characteristic heat in a barrier-overcoming process determines the ability of the heat pump and Onsager's reciprocity coefficients. Then, we give a theoretical derivation of the reciprocity coefficients and show the generality of the results in a class of Feynman's ratchet [5]. We present a simple and clear picture for the mechanism of heat pumps, which is expected to be useful in designing various mesoscopic heat pumps.

Model. – We study a class of systems including Feynman's ratchet in which a particle moves along a periodic structure. The particle interacts with the periodic structure in an asymmetric manner. The periodic structure and the particle are attached to distinct heat baths B_1 and B_2, whose temperatures are T_1 and T_2, respectively. We start from a specific but typical model shown in Fig. 1 [6], which is suitable for numerical simulation. In this

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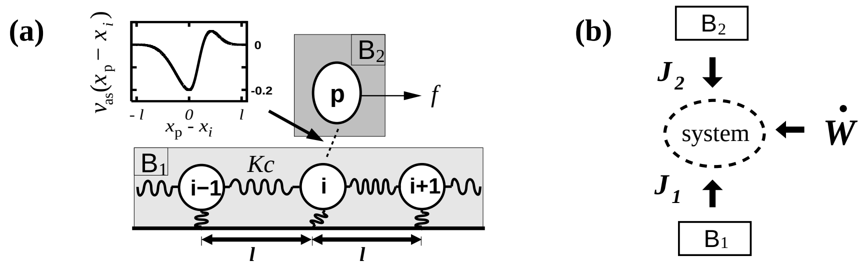

Fig. 1 – (a) Profile of the model for numerical simulation. We fix $\gamma = 1$, $N_\mathrm{c} = 7$ with a periodic boundary condition, and $K_\mathrm{c} = 0.5$ except for Fig. 4. The depth of $v_\mathrm{as}(x_\mathrm{p} - x_i)$ is 0.2. (b) The flow of energy in the system. J_1 and J_2 are mean heat flows from heat baths B_1 and B_2 to the system, and $\dot{W}$ is the mechanical work per unit time provided by the external force.

model, a one-dimensional harmonic lattice corresponds to the periodic structure. The time evolution of the system is described by a set of Langevin equations,

$$\ddot{x}_i = -\gamma \dot{x}_i + \sqrt{2\gamma T_1}\, \xi_i(t) - \frac{\partial (V_\mathrm{as} + V_\mathrm{l})}{\partial x_i}, \quad \ddot{x}_\mathrm{p} = -\gamma \dot{x}_\mathrm{p} + \sqrt{2\gamma T_2}\, \xi_\mathrm{p}(t) - \frac{\partial V_\mathrm{as}}{\partial x_\mathrm{p}} + f, \tag{1}$$

where x_p and x_i are the positions of the particle and the i-th lattice site ($i = 1, \cdots, N_\mathrm{c}$). An external force f is applied to the particle. γ is the friction constant and $\xi_\alpha(t)$ is the random force satisfying $\langle \xi_\alpha(t) \rangle = 0$ and $\langle \xi_\alpha(t) \xi_\beta(t') \rangle = \delta_{\alpha,\beta}\delta(t - t')$. The interaction between the particle and the lattice sites is described by the asymmetric potential $V_\mathrm{as} = \sum_i v_\mathrm{as}(x_\mathrm{p} - x_i)$ with v_as as in Fig. 1. $V_\mathrm{l} = \sum_i v_\mathrm{o}(x_i - x_{i0}) + v_\mathrm{c}(x_{i+1} - x_i)$ is the harmonic potential for the lattice, where $v_\mathrm{o}(x) = K_\mathrm{c} x^2/2$ is the on-site potential and $v_\mathrm{c}(x) = K_\mathrm{c}(x - l)^2/4$ is the interaction between neighbors. l is the lattice interval, $x_{i0} = il$ for i-th site, and K_c is a parameter for stiffness. When K_c is sufficiently large, the lattice does not fluctuate and the potential for the particle reduces to a one-dimensional sawtooth-shaped potential.

On this periodic structure, the particle performs stepwise motions with a typical step size l. At sufficiently low temperatures, each stepwise motion occurs stochastically with a dwell time long enough for the particle to lose its memory. In equilibrium, the probabilities of rightward and leftward steps are of course identical. In the following, we concentrate on nonequilibrium steady states maintained by an external force and/or a temperature difference.

Heat flow induced by particle motion and Onsager relation. – Temporal heat flows from the heat baths to the system are defined for each trajectory by using the stochastic energetics [7,8] as

$$\hat{J}_1(t) = \sum_i [-\gamma \dot{x}_i + \sqrt{2\gamma T_1}\, \xi_i(t)] \circ \dot{x}_i, \qquad \hat{J}_2(t) = [-\gamma \dot{x}_\mathrm{p} + \sqrt{2\gamma T_2}\, \xi_\mathrm{p}(t)] \circ \dot{x}_\mathrm{p}, \tag{2}$$

where $\hat{J}_1(t)$ is the heat flow from B_1 to the system and $\hat{J}_2(t)$ is that from B_2 to the system. From Eq.(2), we define temporal heat flow between the two heat baths as

$$\hat{J}_\mathrm{q}(t) \equiv (\hat{J}_2(t) - \hat{J}_1(t))/2. \tag{3}$$

In the steady states, mean heat flows $J_1 \equiv \langle \hat{J}_1(t) \rangle$, $J_2 \equiv \langle \hat{J}_2(t) \rangle$ and $J_\mathrm{q} \equiv \langle \hat{J}_\mathrm{q}(t) \rangle$ can be defined, where $\langle \cdot \rangle$ denotes average over time and ensemble.

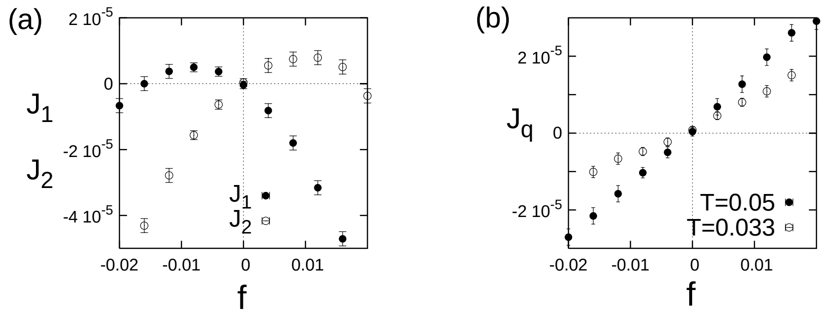

Fig. 2 – (a) Mean heat flows J_1 and J_2 as a function of f at $T_1 = T_2 = 0.05$. For small f, either J_1 or J_2 is positive, which indicates that the system absorbs energy from one of the baths. (b) J_q vs f for $T_1 = T_2 = 0.05$ and 0.033, where $J_\mathrm{q} \propto f$.

When a steady external force f is applied to the particle, energy corresponding to the mechanical work by f is provided to the system. Its mean rate is

$$\dot{W} \equiv \langle \dot{x}_\mathrm{p} f \rangle = J_\mathrm{p} f, \tag{4}$$

where J_p is the mean particle flow defined as $J_\mathrm{p} \equiv \langle \dot{x}_\mathrm{p} \rangle$. In the steady states, the energy of the system remains constant on average. This constraint leads to the balance of the energy flows $\dot{W}$, J_1 and J_2 as in Fig. 1(b), and implies $\dot{W} = -(J_1 + J_2)$.

Let $T_1 = T_2$. When the particle is dragged, the particle and the sites are expected to warm up due to the friction. Then one expects that heat transfer from the hot components to their surroundings takes place, and that $J_1 < 0$ and $J_2 < 0$. On the contrary to this expectation, J_1 or J_2 can be positive for small values of f in Fig. 2(a) ([1]). The system then works as a heat pump which absorbs energy from one of the heat baths and dissipates it to the other.

Fig. 2(b) shows that the heat flow J_q depends linearly on f in a wide range, including the region with $J_1 J_2 > 0$ (see Fig. 2(a)). The particle flow J_p also shows a linear dependence on f in a comparable range. Since the range in which the system works as a heat pump is included in this linear range, we are led to study Onsager relation [9] for J_p and J_q. Define T and ΔT by $T_1 = T - \Delta T/2$ and $T_2 = T + \Delta T/2$. Thermodynamic forces for the flows J_p and J_q are f/T and $\Delta T/T^2$, respectively. Onsager relations are formally written as,

$$J_\mathrm{p} = L_\mathrm{pp}\frac{f}{T} + L_\mathrm{pq}\frac{\Delta T}{T^2}, \qquad J_\mathrm{q} = L_\mathrm{qp}\frac{f}{T} + L_\mathrm{qq}\frac{\Delta T}{T^2}. \tag{5}$$

The coefficients are written as

$$L_\mathrm{pp} = D_\mathrm{p}, \quad L_\mathrm{qq} = T^2\lambda, \quad L_\mathrm{qp} = L_\mathrm{pq} = q^\mathrm{eq} D_\mathrm{p}/l, \tag{6}$$

where D_p is the diffusion constant for the particle in equilibrium, $\lambda \equiv \partial J_\mathrm{q}/\partial \Delta T$ is the heat conductivity of the system and q^eq is the characteristic heat which will be discussed in the next section. To derive the expression for L_pp, we define the mobility as $\mu \equiv \partial J_\mathrm{p}/\partial f$. Then

([1])If the system is not in overdamped limit, the mechanical force can make some part of the system cool down. When $J_2 > 0$, kinetic temperature of the particle is lower than T_2 and, when $J_1 > 0$, that for the lattice sites lower than T_1.

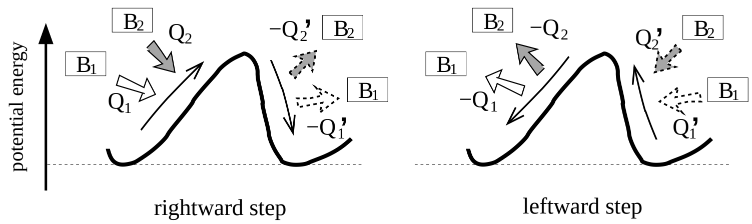

Fig. 3 – Schematics of energy flow when the system overcomes an energy barrier in equilibrium. The bright and dark arrows indicate the heat transfer from the heat bath B_1 and B_2, respectively. In the rightward step, Q_1 is transfered from B_1 during the uphill and $-Q_1'$ during the downhill. Because of the time-reversal symmetry in equilibrium states, Q_1' is transfered from B_1 during the uphill and $-Q_1$ during the downhill in the leftward step. Similar relations hold for B_2 and the relation $Q_1+Q_2=Q_1'+Q_2'$ holds. Because the system has an asymmetry, $Q_1 \neq Q_1'$ and $Q_2 \neq Q_2'$ in general.

Einstein's relation $\mu = D_\mathrm{p}/T$ implies $L_\mathrm{pp} = \mu T = D_\mathrm{p}$. The expression for L_qq follows from the definition. The expressions for the off-diagonal coefficients L_qp and L_pq will be derived in the following (see (7) and (10)).

Characteristic heat associated with barrier-overcoming process. – In this section, we investigate a characteristic heat associated with a barrier-overcoming process in the vicinity of the equilibrium states. It enables us to determine the non-diagonal coefficients L_qp and L_pq. First, let us consider equilibrium cases. When the system exhibits a stepwise motion, it should overcome an energy barrier. To reach the top of the barrier the system absorbs some amount of heat from the two heat baths and dissipates the same amount of heat to descend from the top (Fig. 3). In equilibrium, of course, no net heat transfer occurs between the baths on average. But if we take the ensemble averages of the heat transfer for the individual steps classifying the rightward and the leftward steps, nonvanishing heat transfer between the two baths can survive. We find that a single rightward step carries heat q_R and a leftward step carries q_L on average. In equilibrium, the rightward and the leftward stepwise motions occur with the equal probability and the relation $q_\mathrm{R} = -q_\mathrm{L} \equiv q^\mathrm{eq}$ holds, which consistently leads to vanishing of net heat transfer between the two heat baths. In general, nonvanishing values of q^eq are obtained [10], because it comes from left-right asymmetry of the system.

Next, consider the case where a small external force f is applied under the iso-thermal condition $\Delta T = 0$. From the measurement of J_p and J_q, we can define a characteristic heat $q_\mathrm{c} \equiv J_\mathrm{q}/(J_\mathrm{p}/l)$ in the linear region of J_p and J_q. Because J_p/l is the net number of directed steps per unit time, q_c is conjectured to be a heat transfered per single directed step. By determining the value of q_c and q^eq separately from numerical simulation, we have observed $q_\mathrm{c} \simeq q^\mathrm{eq}$ for sufficiently small f (see Fig. 4(a)). This implies that the characteristic heat q_c associated with the stepwise motion in the linear region is the characteristic heat q^eq at the equilibrium states. Then, we obtain

$$J_\mathrm{q} = \frac{\mu q^\mathrm{eq}}{l} f = \frac{q^\mathrm{eq} D_\mathrm{p}}{l} \frac{f}{T}, \tag{7}$$

by substituting $J_\mathrm{p} = \mu f$ into $J_\mathrm{q}/(J_\mathrm{p}/l) = q^\mathrm{eq}$. Eq. (7) holds for a wide range of the model parameter K_c as is examined in Fig. 4(b), where $\chi_\mathrm{q} \equiv J_\mathrm{q}/f$. Thus the non-diagonal coefficient L_qp is obtained as $q^\mathrm{eq} D_\mathrm{p}/l$. In addition, another non-diagonal coefficient L_pq brings the same

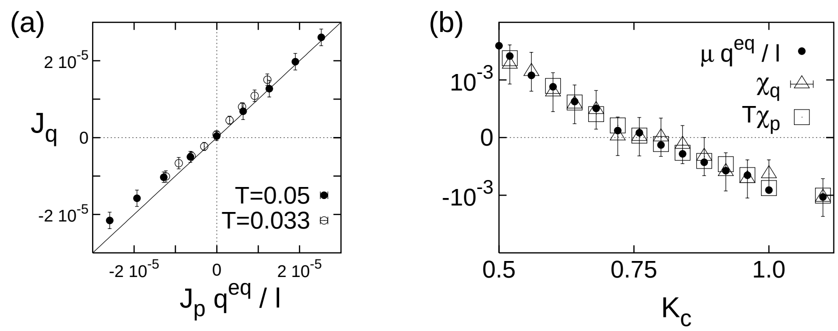

Fig. 4 – (a) Replot of the data in Fig.2(b). Both data for $T = 0.05$ and 0.033 are scaled to the same form as $J_\mathrm{q} \simeq q^\mathrm{eq} J_\mathrm{p}/l$, which corresponds to $q_\mathrm{c} \simeq q^\mathrm{eq}$. J_q and J_p are calculated for $f \neq 0$ while q^eq is evaluated for $f = 0$. (b) The agreement of χ_q with $\mu q^\mathrm{eq}/l$ over various K_c. $T = 0.05$. $\chi_\mathrm{q} \equiv J_\mathrm{q}/f$ and $\mu \equiv J_\mathrm{p}/f$ for small values of f with $\Delta T = 0$. q^eq is evaluated for $f = 0$ and $\Delta T = 0$. At the same time $T\chi_\mathrm{p}$ is plotted, where $\chi_\mathrm{p} \equiv J_\mathrm{p}/\Delta T$ for small values of ΔT with $f = 0$. The observed equality $\chi_\mathrm{q} = T\chi_\mathrm{p}$ indicates that the reciprocity relation $L_\mathrm{qp} = L_\mathrm{pq}$ holds.

form with L_qp via the agreement $\chi_\mathrm{q} = T\chi_\mathrm{p}$ in Fig. 4(b), where $\chi_\mathrm{p} \equiv J_\mathrm{p}/\Delta T$ for small ΔT with $f = 0$. We can rewrite the reciprocity coefficients as $L_\mathrm{qp} = L_\mathrm{pq} = q^\mathrm{eq}\kappa^\mathrm{eq} l$ using the rate constant κ^eq for the barrier-overcoming process of the stepwise motion in equilibrium, where $D_\mathrm{p}=\kappa^\mathrm{eq} l^2$. This form clearly demonstrates that the characteristic heat brought by the barrier-overcoming process results in the reciprocity coefficients.

Generality of the form for reciprocity coefficients. – In this section, we show that the expressions (6) of the reciprocity coefficients are valid in a class of Feynman's ratchet attached to two heat baths. In this class of systems, we can generally define particle flow J_p and heat flow J_q between the two baths and those temporal quantities $\dot{x}_\mathrm{p}(t)$ and $\hat{J}_\mathrm{q}(t)$. The reciprocity coefficients are written with the time correlation function in equilibrium states [11] as

$$L_\mathrm{pq} = L_\mathrm{qp} = \lim_{t_0\to\infty} \frac{1}{2t_0} \int_0^{t_0} \mathrm{d}t \int_{-\infty}^{\infty} \mathrm{d}t' \ \langle \dot{x}_\mathrm{p}(t) \hat{J}_\mathrm{q}(t+t') \rangle \tag{8}$$

where $\langle\cdot\rangle$ denotes ensemble average. Nonvanishing contribution in the r.h.s. of Eq. (8) comes from the stepwise motion of the particle. Supposing that the correlation decays within a finite time and that each stepwise motion is sufficiently separated in time, Eq. (8) is rewritten as a summation of the contribution from each stepwise motion at $t = t_{i_\mathrm{s}}$ as

$$\lim_{t_0\to\infty} \frac{1}{2t_0} \sum_{\{i_\mathrm{s}|t_{i_\mathrm{s}}\in(0,t_0)\}} \int_{t_{i_\mathrm{s}}-\tau}^{t_{i_\mathrm{s}}+\tau} \mathrm{d}t \ \langle \dot{x}_\mathrm{p}(t) \int_{-\tau}^{\tau} \mathrm{d}t' \ \hat{J}_\mathrm{q}(t+t') \rangle, \tag{9}$$

where i_s is the index of the stepwise motion and τ is a time sufficiently longer than the correlation time. Because the integration $\hat{q}_{i_\mathrm{s}} \equiv \int_{-\tau}^{\tau} \mathrm{d}t' \hat{J}_\mathrm{q}(t+t')$ has no explicit dependence on t, the integration of $\dot{x}_\mathrm{p}$ with t can be done separately, which gives $+l$ and $-l$ for rightward and leftward stepwise motion, respectively. Classifying the stepwise motion indexed by i_s into

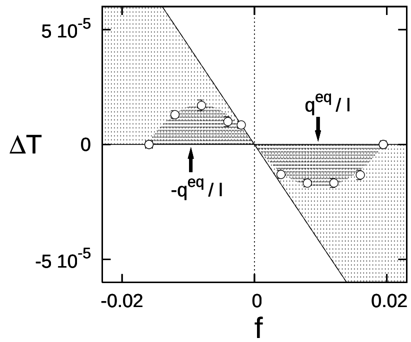

Fig. 5 – The dark shaded area shows the range where the system functions as a heat pump when $T = 0.05$. Numerically identified boundaries of the second inequality of (11) are depicted by (∘). The bright shaded area is obtained from Eq. (12), which is the approximation valid in the linear response regime.

the rightward one indexed by $i_{\rm R}$ and the leftward one indexed by $i_{\rm L}$, Eq. (9) gives

$$L_{\rm pq}=\lim_{t_0\to\infty}\frac{l}{2t_0}\left(\sum_{\{i_{\rm R}|t_{i_{\rm R}}\in(0,t_0)\}}\langle\hat{q}_{i_{\rm R}}\rangle-\sum_{\{i_{\rm L}|t_{i_{\rm L}}\in(0,t_0)\}}\langle\hat{q}_{i_{\rm L}}\rangle\right)=\frac{l}{2}\left(\kappa_{\rm R}\langle\hat{q}_{i_{\rm R}}\rangle-\kappa_{\rm L}\langle\hat{q}_{i_{\rm L}}\rangle\right)=q^{\rm eq}\kappa^{\rm eq}\,l \quad (10)$$

where $\langle\hat{q}_{i_{\rm R}}\rangle = q_{\rm R} = q^{\rm eq}$ and $\langle\hat{q}_{i_{\rm L}}\rangle = q_{\rm L} = -q^{\rm eq}$ [10]. $\kappa_{\rm R}$ and $\kappa_{\rm L}$ are rate constants for the rightward and the leftward stepwise motion respectively and in equilibrium $\kappa_{\rm R} = \kappa_{\rm L} = \kappa^{\rm eq}$. The expression of $L_{\rm pq}$ in Eq. (10) is equivalent to Eq. (6). Furthermore, direct numerical calculations of the time integral of Eq. (8) shows a good agreement with $q^{\rm eq}D_{\rm p}/l$ (data not shown).

Qualification of the heat pump. – A system functions as a heat pump if it can absorb energy from a cooler heat bath and dissipate it to a hotter one. The present system can function as a heat pump when the conditions

$$\Delta T J_{\rm q} \le 0, \quad |J_{\rm q}| \ge \dot{W}/2 \ge 0 \quad (11)$$

are valid. The former inequality means that the heat flow $J_{\rm q}$ between the two heat baths is against temperature gradient ΔT. The latter means that energy is absorbed from one of the heat bath while dissipated to the other, i.e., $J_1 J_2 \le 0$ where $J_1 = -J_{\rm q} - \dot{W}/2$ and $J_2 = J_{\rm q} - \dot{W}/2$.

For the linear response region with small f and ΔT, the conditions (11) are reduced to

$$\Delta T(\Delta T + \mu q^{\rm eq} f/\lambda l) \le 0, \quad (12)$$

which is satisfied in the bright shaded area in f-ΔT plane of Fig. 5. Here we substituted $J_{\rm q}$ of Eq. (5) and (6) into the first inequality of (11). $\dot{W}$ is vanishing in the linear order of f and ΔT because $\dot{W} = J_p f$, which means that the second inequality in (11) always holds.

For larger values of f and ΔT, nonlinear effects (e.g. nonvanishing $\dot{W}$) are not negligible and the region where the system can work as a heat pump is reduced. For instance, for large

values of f with $\Delta T = 0$, we obtain $J_1 < 0$ and $J_2 < 0$ as is seen in Fig. 2(a) although Eq. (12) is satisfied. The region of f and ΔT satisfying the conditions (11) where the system actually functions as a heat pump are numerically identified in the model of Fig. 1(a) as the dark shaded area in Fig. 5. Adjusting the strength of the applied force within this confined area of parameters, we can choose the energy absorption rate from a cooler heat bath. The rate is maximum at $|f| \simeq |q^{\rm eq}|/l$ in the present system (refer Fig. 5) (^{2}). The direction of the heat transfer can also be controlled by the direction of the applied force. The rightward mechanical force induces the absorption of heat from B_2 and the dissipation to B_1 (i.e. $J_1 \leq 0$ and $J_2 \geq 0$), and vice versa for the leftward force ($J_1 \geq 0$ and $J_2 \leq 0$).

With this heat pump, we can cool an object to the temperature limited by the conditions (11) optimizing the applied force. If we need to achieve lower temperature, we should construct a proper structure combining multiple pumps.

Discussion. – In this paper, we have shown that heat transfer can be controlled by a mechanical force. We clarified the conditions for the mesoscopic system to function as a heat pump based on the study of the linear response regime and numerical simulations in fully nonequilibrium. These results are consistent with the results obtained in a discretized solvable model for Feynman's ratchet [4]. The Langevin dynamics adopted in this study suggests an intuitive picture based on the barrier overcoming processes and associated heat transfer $q^{\rm eq}$. The form of the reciprocity coefficients in Eq. (10) generally applies to the class of systems with an asymmetric periodic structure and two heat baths. When the direct measurement of $q^{\rm eq}$ is difficult, we can estimate $q^{\rm eq}$ from the Onsager coefficients as $q^{\rm eq}/l = L_{\rm qp}/L_{\rm pp}$. This estimation can be applied to other models of heat pump, e.g. [4] and [12].

The form of the reciprocity coefficient $L_{\rm qp} = q^{\rm eq}\kappa^{\rm eq} l$ implies that the present heat pump works effectively in small energy scales where barrier-overcoming processes are governed by thermal fluctuations. This is typically realized in molecular scales. The pump is not efficient in very low temperatures where the barrier-overcoming event rarely occurs. It would also be less effective for very high temperatures. This is because the value of $q^{\rm eq}$, which depends on the left-right asymmetry of the system, would be smaller in higher temperatures.

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Note: After the completion of the present work, [13] and [14] were published and [12] appeared in the archive, where different models of heat pump are discussed.

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(^{2})In the present model, Eq. (5) and $\dot{W} = J_{\rm p} f$ give a good estimation of J_1 and J_2, which result in maximum energy absorption rate from a cooler heat bath at $|f| = |q^{\rm eq}|/l(1 + |\Delta T|/2T) \simeq |q^{\rm eq}|/l = L_{\rm qp}/L_{\rm pp}$.

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